

Magnons and electromagnons in a spin-lattice-coupled frustrated magnet CuFeO_2 as seen via inelastic neutron scattering

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We have investigated spin-wave excitations in a four-sublattice (4SL) magnetic ground state of a frustrated magnet CuFeO_2 , in which “electromagnon” (electric-field-active magnon) excitation has been discovered by recent terahertz time-domain spectroscopy [Seki *et al.*, *Phys. Rev. Lett.* **105**, 097207 (2010)]. In a previous study, we identified two spin-wave branches in the 4SL phase by means of inelastic neutron scattering measurements under applied uniaxial pressure [T. Nakajima *et al.*, *J. Phys. Soc. Jpn.* **80**, 014714 (2011)]. In the present study, we have performed high-energy-resolution inelastic neutron-scattering measurements in the 4SL phase, resolving fine structures of the lower-energy spin-wave branch near the zone center. Taking account of the spin-driven lattice distortions in the 4SL phase, we have developed a model Hamiltonian to describe the spin-wave excitations. The determined Hamiltonian parameters have successfully reproduced the spin-wave dispersion relations and intensity maps obtained in the inelastic neutron-scattering measurements. The results of the spin-wave analysis have also revealed physical pictures of the magnon and electromagnon modes in the 4SL phase, suggesting that collinear and noncollinear characters of the two spin-wave modes are the keys to understanding the dynamical coupling between the spins and electric dipole moments in this system.

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I. INTRODUCTION

A triangular lattice antiferromagnet CuFeO_2 (CFO) has recently attracted increasing attention due to its cross-correlated phenomena arising from spin frustration. CFO has a delafossite structure, in which triangular lattice layers of magnetic Fe^{3+} ions are separated by nonmagnetic $\text{O}^{2-}\text{-Cu}^+\text{-O}^{2-}$ dumbbells. From the electronic state of Fe^{3+} ions ($S = 5/2$, $L = 0$) and antiferromagnetic interactions between them, one might expect a noncollinear three-sublattice magnetic ground state, a so-called “120° structure.”¹ However, CFO exhibits, in a ground state, a collinear four-sublattice (4SL) antiferromagnetic order in which the magnetic moments of Fe^{3+} ions are confined along the c axis² [see Fig. 1(a)]. Recent theoretical studies have pointed out that in frustrated magnets, collinear magnetic orderings can be stabilized by strong spin-lattice coupling.^{3,4} Actually, synchrotron radiation x-ray diffraction studies by Terada *et al.*⁵ and Ye *et al.*⁶ have revealed that CFO exhibits “spin-driven” crystal structural transitions; while the crystal structure of CFO is a trigonal structure (space group $R\bar{3}m$) in the paramagnetic (PM) phase, it becomes a monoclinic structure in the 4SL phase. This implies that the lattice degree of freedom is the key to stabilizing the 4SL magnetic order.

CFO is also known as a spin-driven magnetoelectric (ME) multiferroic,⁷ in which a screw-type magnetic order induced by nonmagnetic substitution or application of magnetic

field breaks inversion symmetry of the system and triggers ferroelectricity.^{8–14} Furthermore, recent terahertz time-domain spectroscopy has discovered “electromagnon” (electric-field-active magnon) excitation in the *nonferroelectric* 4SL phase,¹⁵ indicating a dynamical coupling between spins and electric dipole moments in this system.

To understand these various cross-correlated phenomena, it is fundamental to determine the magnetic interaction parameters of this system. Ye *et al.* have performed inelastic neutron-scattering measurements to determine these parameters from spin-wave dispersion relations in the 4SL phase.¹⁶ However, in the previous work, there is some ambiguity in the identification of the spin-wave branches because of a problem arising from magnetic domain structures in this system. Specifically, in the 4SL phase, CFO has three types of magnetic domains whose magnetic propagation wave vectors are described as $(\frac{1}{4}, \frac{1}{4}, \frac{3}{2})$, $(-\frac{1}{2}, \frac{1}{4}, \frac{3}{2})$, and $(\frac{1}{4}, -\frac{1}{2}, \frac{3}{2})$ using a hexagonal basis. These wave vectors are crystallographically equivalent to each other because of the threefold rotational symmetry of the original trigonal crystal structure. Therefore, the magnetic excitation spectrum reported in the previous study¹⁶ is a mixture of spin-wave spectra corresponding to the three different orientations of the magnetic domains.

Quite recently, we have demonstrated that a “single-domain” 4SL phase can be realized by uniaxial pressure applied perpendicular to the c axis.¹⁷ We performed inelastic neutron-scattering measurements under applied uniaxial

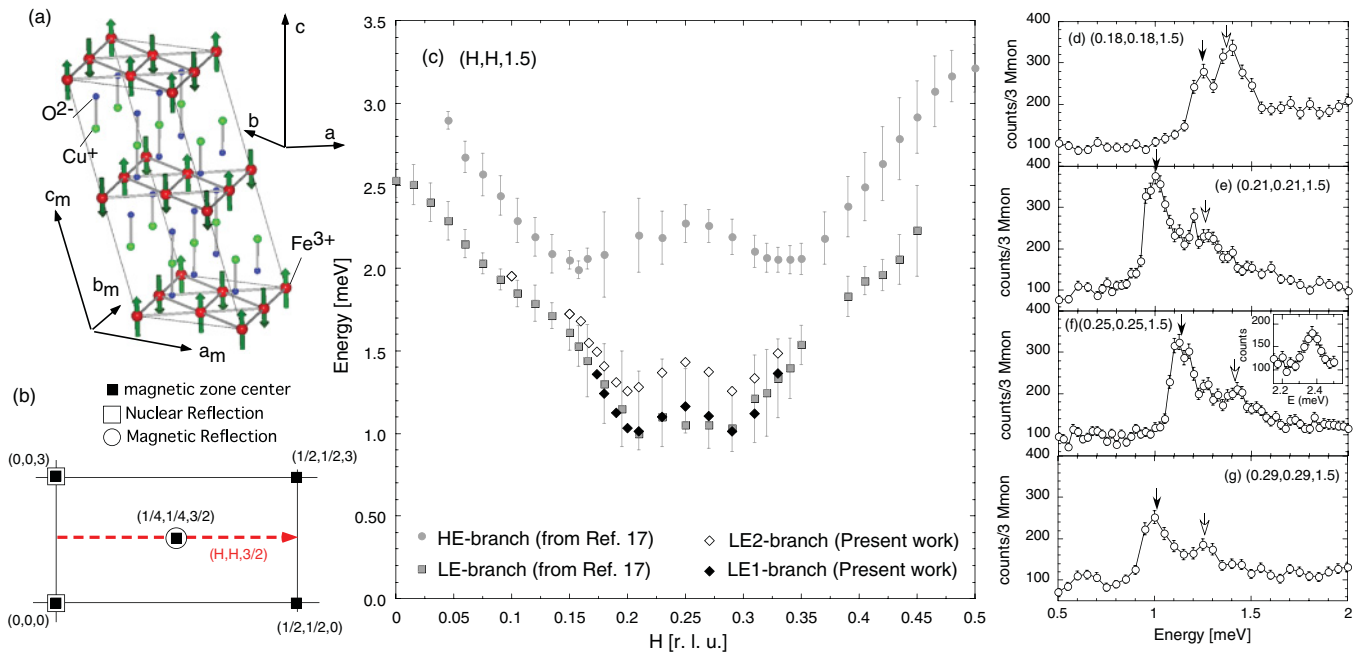


FIG. 1. (Color online) (a) A magnetic unit cell of the 4SL magnetic structure. a_m , b_m , and c_m denote the monoclinic basis. (b) The reciprocal lattice map of the (H, H, L) scattering plane in CFO. (c) The peak positions of the constant- Q scans combined with the results in Ref. 17. The horizontal bars denote full widths at half maximums of the scattering profiles in the previous study. The scattering profiles of the constant- Q scans at (d) (0.18, 0.18, 1.5), (e) (0.21, 0.21, 1.5), (f) (0.25, 0.25, 1.5), and (g) (0.29, 0.29, 1.5) at $T = 2$ K in the 4SL phase.

pressure and identified two spin-wave branches in the single-domain 4SL phase. We have also pointed out that the lower-energy branch splits into two branches near the zone center. However, the wave-vector (Q) dependence of the splitting was not fully investigated.

In the present study, we have thus performed high-energy-resolution inelastic neutron-scattering measurements to resolve the fine structure of the spin-wave dispersion relations in the 4SL phase. Taking into account the spin-driven lattice distortion, we have developed a model Hamiltonian to describe the spin-wave spectra. The results of the spin-wave calculation have revealed the microscopic picture of the magnon and electromagnon modes in the 4SL phase.

II. EXPERIMENT AND RESULTS

We used the single-crystal CFO sample identical to the sample used in Ref. 17. The sample has been set in a uniaxial pressure cell developed by Aso *et al.*¹⁸ A uniaxial pressure of 10 MPa has been applied to the $[1\bar{1}0]$ surfaces of the sample in order to produce the single-domain 4SL state in which the magnetic domains with the wave vector of $(\frac{1}{4}, \frac{1}{4}, \frac{3}{2})$ dominate over the others.¹⁷

We used a cold-neutron triple-axis spectrometer LTAS (C2-1) installed at JRR-3 at the Japan Atomic Energy Agency, Tokai, Japan. The energy of the scattered neutrons was fixed at $E_f = 2.5$ meV. A horizontal focusing analyzer was employed. Energy resolution at the elastic position was 0.06 meV (full width at half maximum). The higher-order contaminations were removed by a cooled Be-filter placed in front of the sample and a room-temperature Be filter placed in front of the analyzer.

As mentioned in the Introduction, we have already identified the two spin-wave branches in our previous work.¹⁷ We have referred to the higher- and lower-energy branches as the HE and LE branches, respectively. To resolve the fine structure of these branches, we performed constant- Q scans at several reciprocal lattice points at $T = 2$ K in the 4SL phase, as shown in Figs. 1(d)–1(g). We found that the scattering profiles apparently have two-peak structures around the energies corresponding to the LE branch. On the other hand, splitting of the HE branch was not observed within the present experimental resolution, as shown in the inset of Fig. 1(f). Therefore, we focused on the LE branch and systematically carried out constant- Q scans along the $(H, H, \frac{3}{2})$ line in the range of $0.15 < H < 0.35$ and $0 < E < 2.0$ meV. In Fig. 1(c), we have plotted the peak positions of the observed scattering profiles together with the results in Ref. 17, revealing the dispersion curves of the two split branches. Hereafter, we refer to the split branches having lower and higher energies as the LE1 and LE2 branches, respectively.

III. CALCULATIONS AND DISCUSSIONS

A. Model Hamiltonian for the spin-wave excitations in the 4SL phase

The first calculation for the spin-wave dispersion relations in the 4SL phase has been presented by Fishman.¹⁹ Although the calculation, in which three in-plane exchange interactions J_1, J_2 , and J_3 , an exchange interaction between adjacent layers J_z , and a uniaxial single-ion anisotropy D are employed, successfully reproduces the dispersion relation of the LE branch, it does not reproduce that of the HE branch. Specifically, in Ref. 19, the excitation bandwidth for the HE branch along

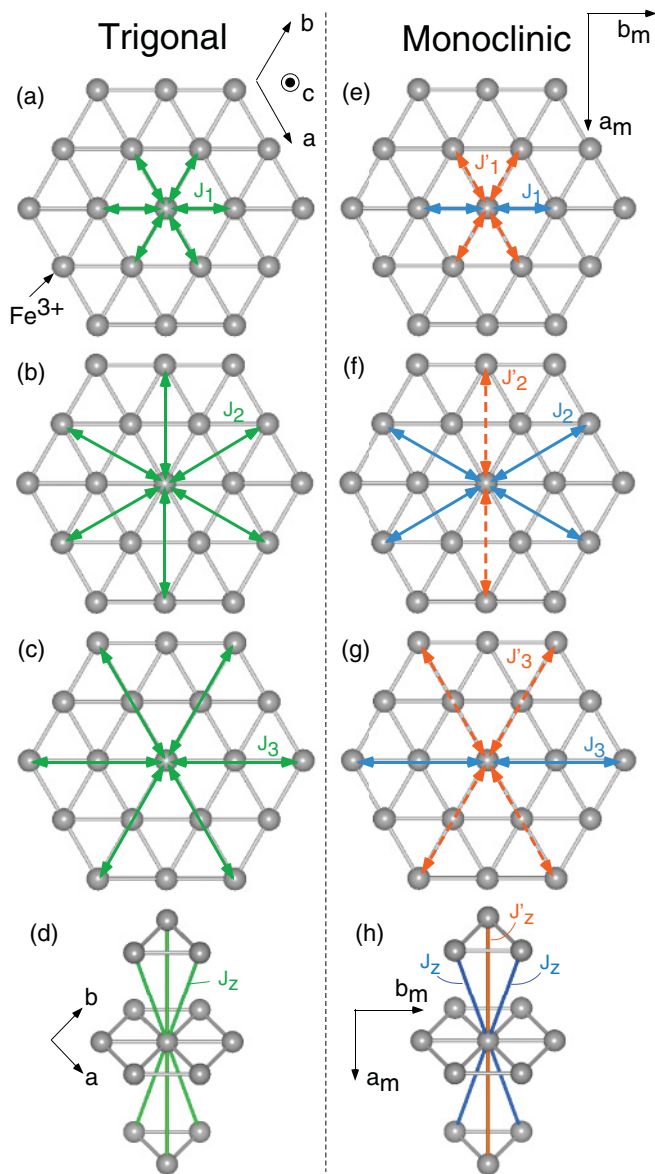


FIG. 2. (Color online) Paths of the exchange interactions (a)–(d) in the trigonal symmetry and (e)–(h) in the monoclinic symmetry of the crystal structure of CFO.

the $(H, H, 3/2)$ line is calculated to be ~ 2.5 meV (from 2.5 to 5.0 meV), while it has been determined to be ~ 1.2 meV (from 2.0 to 3.2 meV) in our experiment.¹⁷ In subsequent study, Fishman *et al.* have corrected the paths of the exchange interactions between the adjacent layers and have presented revised calculations for the spin-wave dispersion relations.²⁰ However, the dispersion relation of the HE branch has not been discussed. These previous calculations^{19,20} are based on the assumption that all of the exchange interactions are isotropic as shown in Figs. 2(a)–2(d). In other words, effects of the spin-driven lattice distortions on the exchange interactions are not taken into account.

On the other hand, Kimura *et al.* have recently discussed the effects of lattice distortions on electron spin resonance (ESR) signals, i.e., spin-wave energies at the zone center, in the 4SL phase.²¹ They have performed multifrequency ESR

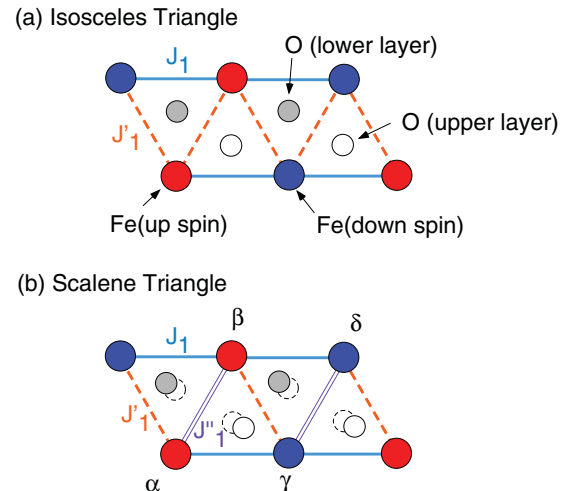


FIG. 3. (Color online) (a) Isosceles and (b) scalene triangle lattices with the 4SL magnetic order.

measurements in the 4SL phase and have analyzed the results using the “scalene triangle model” proposed in the synchrotron radiation x-ray diffraction study by Terada *et al.*⁵ In this model, the nearest-neighbor exchange interaction J_1 splits into three inequivalent interactions due to the monoclinic lattice distortion and displacements of the O^{2-} ions, as shown in Fig. 3(b). These deformations are expected to enhance the antiferromagnetic exchange interactions between neighboring up and down spins and, on the contrary, to reduce those between neighboring two up (or down) spins, as was discussed in Refs. 5 and 22. Consequently, the splitting of J_1 contributes to stabilizing the 4SL magnetic ordering.

Quite recently, Kimura *et al.* have calculated spin-wave dispersion relations in the 4SL phase using the scalene triangle model, assuming that J_1 splits into three inequivalent interactions, while J_2 , J_3 , and J_z are isotropic.²³ They have shown that the energy bandwidth of the HE branch along the $(H, H, 3/2)$ line is calculated to be ~ 1.0 meV (from 1.8 to 2.8 meV). In addition, their calculations have qualitatively reproduced overall features of the results of the inelastic neutron-scattering measurements in the multidomain state.¹⁶ However, there still remain some discrepancies between the shapes of the calculated and observed dispersion curves for the single-domain 4SL phase.

In the present study, we start from three in-plane exchange interactions J_1, J_2 , and J_3 and an interlayer exchange interaction J_z . First, we introduce effects of the trigonal to monoclinic structural transition on all of the exchange interactions. As shown in Figs. 2(e)–2(h), the monoclinic symmetry of the crystal allows the exchange interaction J_i ($i = 1, 2, 3, z$) to split into J_i and J'_i . Second, we have applied the scalene triangle model to the nearest-neighbor exchange interactions, and therefore J'_1 splits into J'_1 and J''_1 , as shown in Figs. 3(a) and 3(b). Although the scalene triangle distortion might affect the other distant interactions, we have assumed that it affects only the nearest-neighbor interactions in the present analysis.

As for the single-ion anisotropy, Fishman *et al.* have employed only a uniaxial anisotropy D .^{16,19,20} On the other hand, Kimura *et al.* have argued that an in-plane anisotropy (E term) is necessary to explain the results of their ESR

measurements.²¹ We have thus employed D and E terms in the present analysis. Therefore, we have written a Hamiltonian for the 4SL phase as

$$\begin{aligned} \mathcal{H} = & -\frac{1}{2} \sum_{i,j} \sum_{n=1}^3 J_n \mathbf{S}_i \cdot \mathbf{S}_j - \frac{1}{2} \sum_{i,j} \sum_{n=1}^3 J'_n \mathbf{S}_i \cdot \mathbf{S}_j \\ & - \frac{1}{2} \sum_{i,j} J''_1 \mathbf{S}_i \cdot \mathbf{S}_j - \frac{1}{2} \sum_{i,j} J_z \mathbf{S}_i \cdot \mathbf{S}_j - \frac{1}{2} \sum_{i,j} J'_z \mathbf{S}_i \cdot \mathbf{S}_j \\ & - \sum_i D (\mathbf{S}_i^z)^2 - \sum_i E [(\mathbf{S}_i^x)^2 - (\mathbf{S}_i^y)^2], \end{aligned} \quad (1)$$

where the x, y , and z axes are defined to be parallel to the $[110]$, $[\bar{1}10]$, and $[001]$ directions of the crystal, respectively.

B. Calculation for the spin-wave spectrum

We have calculated the spin-wave energies in the 4SL phase, applying a Holstein-Primakoff $1/S$ expansion about the classical limit to the Hamiltonian of Eq. (1). Following the previous work by Fishman,¹⁹ we refer to the four magnetic sublattices as α, β, γ , and δ , as shown in Fig. 3(b). We express the spins \mathbf{S}_i on the sublattice α (β, γ , and δ) using the boson operators α_i (β_i, γ_i , and δ_i).

The procedures of the calculation for the spin-wave energies are essentially the same as those in Ref. 19. The spin-wave energies at wave vector q , ϵ_q , are obtained by solving the Heisenberg equation of motion for the vector $\mathbf{v}_q = (\alpha_q, \beta_q, \gamma_q^\dagger, \delta_q^\dagger, \alpha_{-q}^\dagger, \beta_{-q}^\dagger, \gamma_{-q}, \delta_{-q})$, where α_q, β_q , and so on are the Fourier-transformed boson operators. The equation of motion for \mathbf{v}_q can be written using the 8×8 matrix $M(q)$ as $i d\mathbf{v}_q/dt = -[H, \mathbf{v}_q] = M(q)\mathbf{v}_q$. Diagonalizing the matrix $M(q)$, we have obtained the spin-wave energies in the 4SL phase.

To find a suitable set of magnetic interaction parameters, we have prepared 5×10^8 sets of the parameters using the Monte Carlo method²⁴ and have calculated spin-wave energies ϵ_{q_i} , where i stands for a observed data point. For each set of the parameters, we have calculated a sum of squared residuals $\chi^2 = \sum_i (E_{q_i} - \epsilon_{q_i})^2$, where E_{q_i} is an observed spin-wave energy, and then have searched a set of the parameters having a small value of χ^2 . It should be noted that, in principle, a finite in-plane anisotropy (E term) splits not only the LE branch but also the HE branch into two branches. Therefore, we have four calculated spin-wave branches referred to as the LE1, LE2, HE1, and HE2 branches. Since we could not observe the splitting of the HE branch in our inelastic neutron-scattering measurements, we have used E_{q_i} of the observed HE branch for both of the calculated HE1 and HE2 branches in the evaluation of χ^2 .

Examining the 5×10^8 sets of the parameters, we have obtained a set of parameters that successfully reproduces the

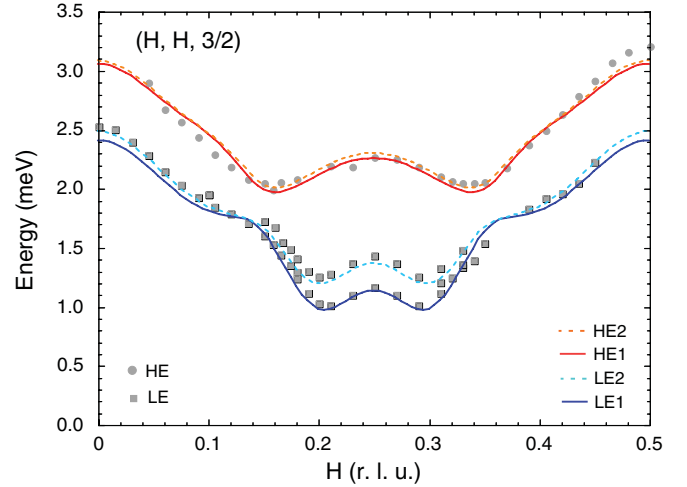


FIG. 4. (Color online) Spin-wave dispersion relations along the $(H, H, \frac{3}{2})$ line calculated with the parameters in Table I and the observed spin-wave energies.

observed dispersion relations. Table I shows the values of the Hamiltonian parameters.²⁵ In Fig. 4, we show the calculated spin-wave dispersion relations along the $(H, H, 3/2)$ line with the observed spin-wave energies.²⁶ Using these parameters, we have also calculated inelastic neutron-scattering cross sections described as

$$\begin{aligned} \frac{d^2\sigma}{d\Omega dE'} \propto & f^2(\kappa) (1 + \hat{\kappa}_z^2) \frac{k_f}{k_i} \sum_{\mathbf{R}, \mathbf{R}'} e^{-i\kappa \cdot (\mathbf{R} - \mathbf{R}')} \\ & \times \int_{-\infty}^{\infty} dt e^{-i\omega t} (\langle S_{\mathbf{R}}^+(0) S_{\mathbf{R}'}^-(t) \rangle + \langle S_{\mathbf{R}}^-(0) S_{\mathbf{R}'}^+(t) \rangle), \end{aligned} \quad (2)$$

where κ is the scattering vector defined as $\kappa = \mathbf{k}_f - \mathbf{k}_i$, \mathbf{k}_f and \mathbf{k}_i are wave vectors of the scattered and incident neutrons, respectively, $f(\kappa)$ is the magnetic form factor of a Fe^{3+} ion,²⁷ κ_z is the z -axis component of a unit vector of the scattering vector, and \mathbf{R} denotes a position of a spin of a Fe^{3+} ions. To obtain resolution-convoluted inelastic neutron-scattering spectra, we have employed the Cooper-Nathans-type resolution function.²⁸ As shown in Figs. 5(a)–5(d), we have demonstrated that the calculated intensity maps show good agreement with the observed data (taken from Ref. 17).

It should be noted that the spin-wave dispersion relations in the (H, H, L) plane does not depend on J'_2 because the direction of J'_2 is perpendicular to the (H, H, L) plane. To determine J'_2 , we have calculated the spin-wave spectra in the magnetically multidomain state. In the magnetic domains having wave vectors of $(-\frac{1}{2}, \frac{1}{4}, \frac{3}{2})$ and $(\frac{1}{4}, -\frac{1}{2}, \frac{3}{2})$ the directions of J'_2 are not perpendicular to the (H, H, L) scattering plane. Hence, the spin-wave spectra belonging to these domains contain information on J'_2 . We have adjusted the value of J'_2

TABLE I. The Hamiltonian parameters (in meV).

$J_1 S$	$J'_1 S$	$J''_1 S$	$J_2 S$	$J'_2 S$	$J_3 S$	$J'_3 S$	$J_z S$	$J'_z S$	DS	ES
-0.455	-0.422	-0.150	-0.100	-0.106	-0.338	-0.372	-0.187	-0.167	0.160	-0.035

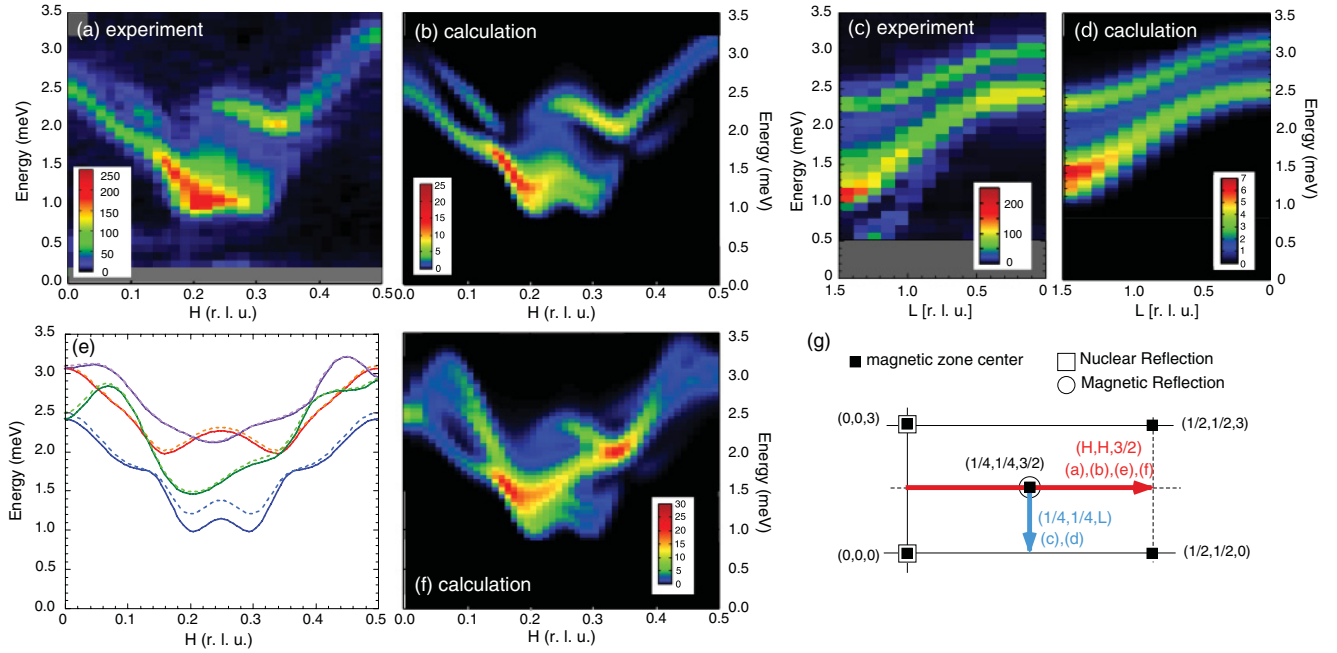


FIG. 5. (Color online) Observed intensity maps of the inelastic neutron-scattering measurements along (a) the $(H, H, 3/2)$ and (c) $(1/4, 1/4, L)$ lines (taken from Ref. 17). Calculated intensity maps along (b) the $(H, H, 3/2)$ and (d) $(1/4, 1/4, L)$ lines for a single-domain 4SL phase. (e) Spin-wave dispersion relations for a magnetically multidomain 4SL phase calculated from the parameters in Table I. (f) Calculated intensity map for the multidomain state in which volume fractions of the three domains are assumed to be equal to each other. (g) The reciprocal lattice map of the (H, H, L) scattering plane. Arrows denote the directions of the $(H, H, 3/2)$ and $(1/4, 1/4, L)$ lines.

so that the calculated multidomain spectrum is close to the observed data in Ref. 16. As a result, $J'_2 S$ is determined to be -0.106 meV. In Figs. 5(e) and 5(f), we show the calculated spin-wave dispersion relations and an intensity map for an inelastic neutron-scattering measurement in the multidomain state in which the volume fractions of the three domains are assumed to be equal to each other, respectively.

We now discuss the determined values of the Hamiltonian parameters. Among the nearest-neighbor exchange interactions, the magnitude of the exchange interactions connecting two up (or down) spins $|J''_1|$ is determined to be smaller than $|J_1|$ and $|J'_1|$, which connect up and down spins. This is consistent with the nature of the scalene triangle distortion; specifically, the nearest-neighbor exchange interactions vary so as to lower the total exchange energy of the 4SL magnetic structure. It is worth mentioning that previous x-ray diffraction studies on CFO^{29} and $\text{CuFe}_{1-x}\text{Al}_x\text{O}_2^{30}$ have suggested that this scalene triangle distortion occurs not only in the 4SL phase but also in the screw-type helimagnetic (multiferroic) phase. Haraldsen *et al.* have recently presented an inelastic neutron-scattering and theoretical study on magnetic excitations in the helimagnetic phase of Ga-doped CFO.³¹ They have assumed that the oxygen displacements also break the equilateral symmetry of J_1 so as to lower the total exchange energy in the helimagnetic phase. Their calculations have successfully reproduced magnetic excitations from the noncollinear magnetic structure. This ensures that the oxygen displacements significantly affect the competing magnetic interactions in this system.

We have also found that the second- and third-neighbor in-plane exchange interactions and interplane interactions are affected by the monoclinic lattice distortion, specifically,

$J'_2/J_2 = 1.06$, $J'_3/J_3 = 1.10$, and $J'_z/J_z = 0.89$. This suggests that the effects of the monoclinic structural transition, which are not taken into account in the previous studies,^{16,19–21} are important to explain the spin-wave excitation in the 4SL phase.

C. Microscopic picture of the electromagnon excitation

As mentioned in the Introduction, Seki *et al.* have recently discovered the electromagnon excitation in the 4SL phase.¹⁵ Specifically, their optical spectroscopy has detected two groups of signals corresponding to the zone-center spin-wave modes belonging to the LE and HE branches. Hereafter, we refer to these spin-wave modes as LE and HE modes, respectively. Measuring these signals with various light polarization configurations, they have concluded that the HE mode is electromagnon driven by an ac electric field, and the LE mode is conventional magnon. To investigate the mechanism of the electromagnon excitation, it is essential to establish physical pictures of these spin-wave modes. In the following discussion, we have neglected the in-plane anisotropy E for simplicity, and therefore there are only two spin-wave modes corresponding to the LE and HE modes because the LE1 and LE2 modes (the HE1 and HE2 modes) degenerate without the E term.

In Figs. 6(a) and 6(b), we have shown the schematic drawings of the LE and HE modes at the zone center, which are obtained by the spin-wave analysis without the E term. These pictures are consistent with the spin-wave modes presented in the recent ESR study by Kimura *et al.*²³ Each spin rotates about the z axis with an angular frequency of $\omega_0^{\text{LE/HE}}$, where

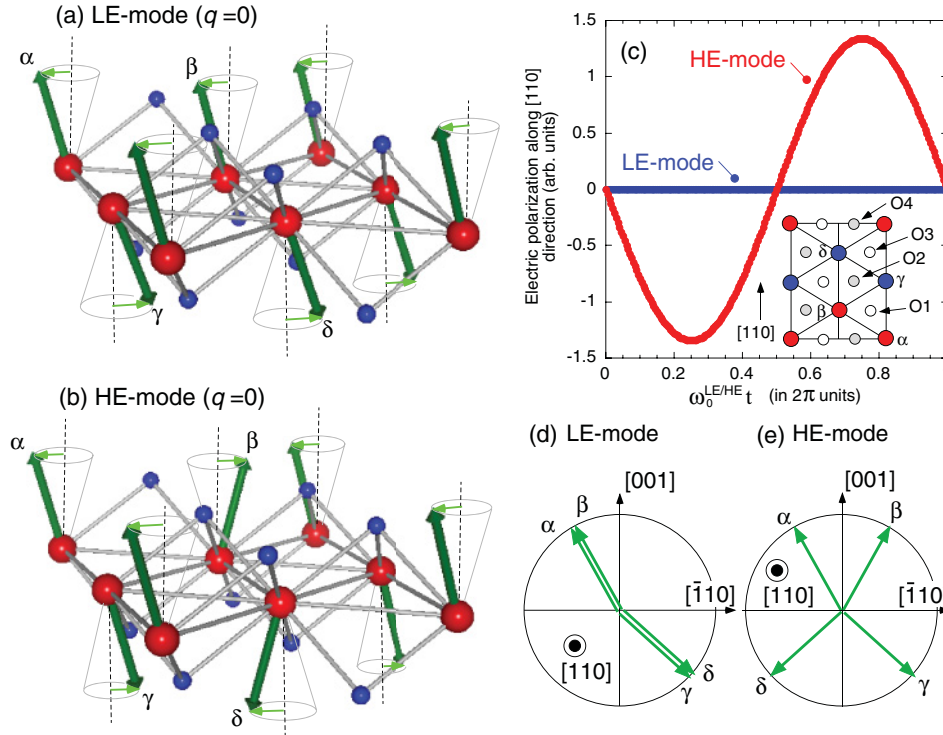


FIG. 6. (Color online) Schematic drawing of the motion of the spins in (a) the LE- and (b) HE-modes. (c) Calculated electric polarization in a magnetic unit cell for the LE- and HE-modes. Inset shows positions of the four inequivalent oxygen sites in a magnetic unit cell. [110]-plane-projections of the spins of the (d) LE- and (e) HE-modes.

$\omega_0^{\text{LE/HE}}$ is the spin-wave frequency of the LE/HE branch at $q = 0$. In the LE mode, the up spins (or the down spins) rotate, keeping a “collinear” configuration. On the other hand, in the HE mode, the two up spins (or two down spins) are coupled in a “noncollinear” configuration in a magnetic unit cell. In Figs. 6(d) and 6(e), we show [110]-plane projections of the spins depicted in Figs. 6(a) and 6(b), respectively. We found that the HE mode has a screwlike spin texture but the LE mode does not, as was predicted in Ref. 15. Moreover, the spin helicity of the screwlike spin texture is reversed by rotating the spins about the z axis by π . Keeping the relationship between the ferroelectricity and the screw-type magnetic ordering in the multiferroic phase of CFO⁷⁻¹⁴ in mind, we anticipate that the noncollinear nature of the HE mode accounts for the dynamical ME coupling in the 4SL phase.

To quantitatively investigate the difference between the LE and HE modes, we have applied, to the two modes, the d - p hybridization model, which successfully explains that the screw-type magnetic ordering in this system can induce ferroelectricity through the variation in the metal-ligand (d - p) hybridization with spin-orbit coupling.³² While this model has been proposed for the relationship between a static magnetic ordering and ferroelectricity, we have employed this model to explain the dynamical ME coupling in the 4SL phase. This model requires three noncollinear spins surrounding an oxygen ion as a minimal unit to produce uniform electric polarization, as was discussed in Ref. 32. Hence, we have calculated a sum of electric polarizations at four inequivalent oxygen sites, each of which is surrounded by three Fe ions in a magnetic unit cell [see the inset in Fig. 6(c)]. The procedures of the calculation are

essentially the same as those in Refs. 30 and 32. Fig. 6(c) shows the calculated electric polarizations along the [110] direction as functions of $\omega_0^{\text{LE/HE}}t$ for the LE and HE modes, revealing that the HE mode generates electric polarization oscillating with the same angular frequency as ω_0^{HE} , but the LE mode does not. This oscillating electric polarization can be coupled with the ac electric fields. We have thus concluded that this must be the microscopic mechanism of the electromagnon excitation in this system.

IV. CONCLUSION

We have investigated spin-wave excitations in the 4SL magnetic ground state of a frustrated magnet CuFeO₂. By high-energy-resolution neutron inelastic scattering measurements, we have revealed the fine structure of the spin-wave dispersion relations in the 4SL phase. Taking account of the trigonal to monoclinic structural transition and the scalene triangle distortion in the 4SL phase, we have developed a model Hamiltonian and have determined the Hamiltonian parameters. We have successfully reproduced the spin-wave dispersion relations and the intensity maps for the inelastic neutron-scattering measurements in the single-domain and multidomain 4SL phase. Consequently, we have concluded that the spin-driven lattice distortions significantly affect the exchange interactions in this system.

Using the results of the spin-wave analysis, we have presented the physical pictures of the zone-center spin-wave modes belonging to the LE and HE branches and

have discussed the mechanism of the recently discovered electromagnon excitation. The pictures of the LE and HE modes suggest that the symmetry of the spin-wave modes are the key to understanding this kind of dynamical ME coupling. Applying the d - p hybridization model³² to the two modes, we have demonstrated that only the HE mode generates the oscillating electric polarization, which can be coupled with an ac electric field, but the LE mode does not. This indicates that the d - p hybridization model, in which spins and electric dipole moments are mediated by a spin-orbit coupling, can explain

not only the static coupling between the magnetic ordering and the ferroelectricity but also dynamical ME coupling, i.e., electromagnon excitation.

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²⁵We could not perform a least-squares analysis because of strong correlations between the Hamiltonian parameters. Therefore, we could not estimate the error bars of the parameters shown in Table I.

²⁶In Ref. 17, we have pointed out that the spin-wave energies at $(1/2, 1/2, 0)$ and $(1/4, 1/4, 3/2)$ are different from each other, although both of the reciprocal lattice points are the zone center. At this moment, the reason for the difference between the spin-wave energies at $(1/2, 1/2, 0)$ and $(1/4, 1/4, 3/2)$ is not clear. In the present analysis, we have determined the Hamiltonian parameters so that the calculated and observed spin-wave energies agree with each other at $(1/4, 1/4, 3/2)$.

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